

Recent trends in investigation of atomic subshell processes accompanying photon-atom interaction

K. L. Allawadhi

Nuclear Science Laboratories, Department of Physics
Punjabi University, Patiala - 147 002, INDIA

Abstract: Atomic subshell processes accompanying photon-atom interactions include the processes responsible for creation of subshell vacancies and their subsequent filling up. The subshell and level-to-level transition parameters relating to these processes include subshell photoionization and Compton scattering cross sections, subshell fluorescence yields, subshell Auger and Coster-Kronig transition probabilities, line intensities and level decay rates, level widths and life times etc. In this paper the status of the calculated and experimental values of these parameters, comparison between the two and the trends and approaches being applied to the investigations are briefly described.

Keywords: subshell processes, inner shells, photon-atom interaction, fluorescent x-rays

PACS numbers: 32.80.-t, 32.80.Fb

1. Introduction

Atomic subshell processes accompanying photon-atom interactions include the processes responsible for creation of subshell vacancies and their subsequent filling up. The processes responsible for the creation of the vacancies following the interaction, especially in the inner shells, include: (a) photoelectric and Compton interaction with subshell electrons, (b) radiative and

non radiative transfer of vacancies from lower shells to higher shells, (c) Coster-Kronig transfer of vacancies from lower subshells to the upper subshells of a same shell and (d) interaction of emitted electrons (photo, Compton and Auger) with subshell electrons, wherever energetically possible. While the first three (a-c) result in vacancies relating to the primary interactions, the last one (d) is secondary in nature.

The vacancies thus created are filled by transitions from the higher shells either through radiative processes or nonradiative processes. The first of these two results in emission of x-rays which may either be emitted as a diagram line (resulting from a single vacancy) or nondiagram line (resulting from multiple or spectator vacancies). The nondiagram lines, thus emitted, are also known as satellite lines. The nonradiative processes include the Auger electron emission (resulting from inter-shell transitions) or Coster-Kronig transfer (resulting from intra-shell transitions).

The subshell and level-to-level transition parameters pertaining to the above processes are: (a) subshell photoionization and Compton scattering cross sections, (b) subshell fluorescence yields, (c) subshell Auger and Coster-Kronig yields, (d) line intensities, (e) level decay rates, level widths and life times etc.

In this paper the status of the calculated and experimental values of these parameters, comparison between the two and the trends and approaches being applied to the investigations are briefly described.

2. Calculations and measurements of different parameters

Theoretical calculation of different parameters relating to subshells and level-to-level transitions, such as subshell photoionization cross sections, subshell x-ray fluorescence, Auger and Coster-Kronig yields, line intensities, level decay rates, life time, level widths, etc., have been made using different assumptions and approximations. In order to limit the size of the present communication only some of the important tabulations / reports are listed below:

- (1) Scofield [1] has calculated photoionization cross-sections for all shells/subshells of atoms with $Z=1-100$ in the energy range 1-1500 keV.
- (2) Hubbell et al. [2] have tabulated the values of incoherent scattering function for $Z=1-100$. These scattering functions can be used to convert free-electron Klein-Nishina cross sections to Compton scattering cross-sections for bound electrons. These tabulations are said to be constructed from available state-of-the-art theoretical data.
- (3) Krause et al. [3] and Chen et al. [4] have generated sets of values of K - and L -shell/subshell fluorescence yields and Coster-Kronig transition

probabilities. While the values of Krause et al. are available for all elements with atomic numbers $5 \leq Z \leq 104$, values of Chen et al. are available for 25 different elements with atomic number ranging from 60 to 100.

- (4) Scofield [5,6] has calculated x-ray emission rates for filling up of the vacancies in the *K* and *L* shell and also presented the total *K*- and *L*-shell radiative decay rates and rates of emission of individual x-ray lines in elements with $Z=5-104$.
- (5) Rao et al. [7] have generated semi-empirical values of the probabilities for shifting a *K* vacancy to *L* subshells for the elements with even atomic number in the range $Z=20-94$.
- (6) Allawadhi et al. [8] have calculated L_1 , L_α , L_β and L_τ x-ray production cross sections in all elements with $40 \leq Z \leq 92$ at all energies between the L_1 edge to just below the *K* edge of these elements.
- (7) Hubbell et al. [9] have tabulated *K*-, *L*-, *M*- and higher-shell atomic x-ray fluorescence yields, fitted to standard empirical parametric formulations.
- (8) Mittal et al. [10] have generated optimum values of average *L* shell fluorescence yields in elements $23 \leq Z \leq 92$.
- (9) Some meager [1,11,12] data on these parameters for the *M* shell is also available in literature.

The calculated values of these parameters are thus available, now, over a wide range of incident photon energies and some of these are available even for almost the entire periodic table. However, the position of the experimental work [8-16] in this direction is not in tune with the calculated values and amounts to the measurements of some gross parameters only. The different parameters measured in photon-atom interaction so far, for example, include:

- (1) *K*- and total *L*-shell photoionization cross section at some selected photon energies,
- (2) K_α , K_β , L_1 , L_α , L_β , L_τ and total *M*-shell x-ray production cross section in some elements and some parameters derived therefrom.
- (3) Measurement of the angular distribution and polarization of photon induced *K*-, *L*- and *M*-shell x-rays in few elements.

3. Comparison between experimental and theoretical values – different approaches

As the extent of experimental data available so far, as mentioned above, is not sufficient to meet the need of these parameters in theory and experiment

in pure and applied sciences, one has to rely largely on the theoretical or semi-empirical data available in literature. For use of the theoretical or semi-empirical values of these parameters in other calculations or experiments, the values must be checked against experiments. However, in most of the cases the direct comparison of theory with experiment is difficult because while theoretical calculations are directly available for subshell and level-to-level transition parameters, the availability of experimental values is still limited to some gross parameters only as mentioned above. The nonavailability of the direct experimental data on subshells and level-to-level transitions is, possibly, because of the following two reasons:

- (1) The first reason is related to the limit of the present day instrumentation. The best resolution available in the energy dispersive x-ray detectors (Si(Li) or HpGe), which have been largely used for such measurements, is of the order of 150 eV at the Mn K_{α} x-ray energy (about 5.9 keV). With this resolution the K and L x-rays can not be resolved in more than 4 to 5 components / groups depending on the Z value of the emitting element. The actual number of lines in the K x-ray and L x-ray spectra of different elements varies from 6-8 for K -shell, and 30-40 for L -shell x-rays. Wavelength dispersive x-ray spectrometers though have much better resolution than that of energy dispersive counterparts, but these have not been largely used for the purpose because of their slow speed and very high cost.
- (2) The second reason for the lack of such experimental data is the presence of faster Coster-Kronig transitions which alter the vacancies within the subshells of the same shell before these are filled by the transitions from the higher shells. Thus, the characteristics studied experimentally are, in general, of the altered vacancies and do not pertain to the primary vacancy distribution.

From the above it is clear that for the comparison between theory and experiment, one has to either modify the theoretical calculations / semi-empirical data on some of the basic parameters so as to obtain the compound gross parameters measured experimentally so that the comparison become possible, or refine the measurements so as to obtain the direct values of sub-shell or level-to-level transitions parameters.

In the first approach, which has been largely followed in the past or even the recent past, the parameters normally calculated for comparison with theory contain large uncertainties, for example, the theoretical values of the L_1 , L_{α} , L_{β} and L_{τ} x-ray production cross-sections, $\sigma_{L\tau}$, are calculated from the theoretical values of L -subshell photoionization cross sections, fluorescence yields w_i and Coster-Kronig transition probabilities, f_{ij} , radiative decay

rates, F_i and intra-shell transition probabilities, n_{KL_i} , using the following relations;

$$\sigma_{L1} = [(\sigma_{L1} + \sigma_K n_{KL1}) (f_{12} f_{23} + f_{13}) \\ + (\sigma_{L2} + \sigma_K n_{KL2}) f_{23} + (\sigma_{L3} + \sigma_K n_{KL3})] w_3 F_{31}$$

$$\sigma_{L\alpha} = [(\sigma_{L1} + \sigma_K n_{KL1}) (f_{12} f_{23} + f_{13}) \\ + (\sigma_{L2} + \sigma_K n_{KL2}) f_{23} + (\sigma_{L3} + \sigma_K n_{KL3})] w_3 F_{3\alpha}$$

$$\sigma_{L\beta} = (\sigma_{L1} + \sigma_K n_{KL1}) w_1 F_{1\beta} \\ + [(\sigma_{L1} + \sigma_K n_{KL1}) f_{12} + (\sigma_{L2} + \sigma_K n_{KL2})] w_2 F_{2\beta} \\ + [(\sigma_{L1} + \sigma_K n_{KL1}) (f_{12} f_{23} + f_{13}) \\ + (\sigma_{L2} + \sigma_K n_{KL2}) f_{23} + (\sigma_{L3} + \sigma_K n_{KL3})] w_3 F_{3\beta}$$

$$\sigma_{L\tau} = (\sigma_{L1} + \sigma_K n_{KL1}) w_1 F_{1\tau} \\ + [(\sigma_{L1} + \sigma_K n_{KL1}) f_{12} + (\sigma_{L2} + \sigma_K n_{KL2})] w_2 F_{2\tau}$$

$K-L_1$ being a forbidden transition, the values of n_{Li} in the expressions above have been taken as:

$$n_{L1} = n_{L1}(A) \\ n_{L2} = n_{L2}(A) + n_{L2}(R) \\ n_{L3} = n_{L3}(A) + n_{L3}(R)$$

where, R or A in the parentheses indicate that the term belongs to a radiative or an Auger transition, respectively.

When the K -shell binding energy of the secondary target element is greater than the energy of the K conversion x-rays of the primary target element, the value of K -shell photoionization cross section σ_K , becomes zero in the above relations.

It is seen that if the uncertainties in the values of these cross sections are estimated, these turn out to be of the order of 20-25% while the claimed accuracy in the available experimental values are of the order of 8-10%. The same applies to some of the other parameters, e.g., the total L -shell fluorescence

yields.

In the second approach, experimental methods have to be devised so as to measure the basic subshell and level-to-level transition parameters directly and, thus, make a direct comparison of the measured value with that of the theoretical value. This approach has not been largely followed for measurements of parameters relating to photon-atom interactions because of the reasons explained above.

4. Conclusions and directions

From the above discussion it is clear that so far as the investigations relating to processes following photon-atom interaction in the coming years are concerned, concentrated and systematic efforts are needed in following directions:

- (1) The extent of accuracy in the theoretical / semi-empirical values of the basic parameters have to be improved so that the gross parameters generated from these have accuracy equal to or even better than that of their experimental counterpart.
- (2) The experimental techniques have to be improved so as to make direct measurements of subshell and level-to-level transitions parameters possible. As this will make the direct comparison between theory and experiment possible.

Some of the areas in which investigations of the subshell processes and the parameters accompanying photon-atom interaction still needed are:

- (1) L_1 -, L_2 - and L_3 -subshell photoionization cross sections. [No experimental data are available.]
- (2) L -subshell Coster-Kronig transition probabilities. [The uncertainties in the scanty available data are large and exceed even 50% in some of the cases.]
- (3) Satellite K - and L -shell line intensities. [The data on satellite lines in parametric form is not available so far.]
- (4) K to $L_1/L_2/L_3$, K to $M_1/M_2/M_3/M_4/M_5$, and $L_1/L_2/L_3$ to $M_1/M_2/M_3/M_4/M_5$ vacancy transfer probabilities. [Experimental data are not available in most of the cases.]
- (5) All basic parameters in case of M and higher shells. [Either no or vary scanty experimental data are available.]

The most important part of the methodology to investigate these parameters is to measure precisely and accurately the energy and intensity of different x-ray lines emitted from the interacting system following the photon-atom

interaction. The techniques which are found to be very useful in the measurements of subshell parameters and for level-to-level transitions include: (a) selective absorption techniques, (b) selective excitation techniques, (c) use of computer spectroscopy software, and (d) coincidence techniques.

In the selective absorption technique, an absorber of a suitable element and thickness having large absorption for photons of one energy but low absorption for the other is used. The technique was tried by the investigators for separation of $L_{\alpha 1}$ and $L_{\alpha 2}$ x-rays of Au using Zn absorber and the angular distribution and polarization [17] of these x-ray lines were investigated.

In the selective excitation technique, the energy of incident photons is selected in such a way that it can excite the electrons in a particular subshell (e.g., the L_3 subshell) but is not sufficient to excite the next immediate shell/subshell (e.g., the L_2 subshell). In this way, the effect of Coster-Kronig transitions can either be completely eliminated or at least minimized. This technique can be applied using photon induced fluorescent K x-rays, Compton-scattered gamma rays and synchrotron radiation. Out of these, the first two have been tried by the investigators [18,19] as follows: (i) the angular distribution and polarization of L_3 subshell x-rays in Th and U have been investigated using Nb and Mo K x-rays, respectively; (ii) the effect of Coster-Kronig transitions on aligned vacancies has been investigated on U using Mo and Sn K x-rays; (iii) a technique for energy and efficiency calibration of a HpGe detector using variable energy Compton-scattered gamma rays has been developed; and (iv) the absorption coefficients in some rare earth elements at energies just below and above K edge has been investigated using variable energy Compton-scattered gamma rays. Synchrotron radiation sources are not yet available in India, however, this technique has also been well explored by some workers [20] wherever these facilities are available.

Computer spectroscopy software is also found to be very useful and is being used to resolve the spectral x-ray peaks which do not otherwise appear separate in the recorded spectrum. This technique has replaced the older graphical methods used for peeling of different peaks. The technique has been tried by the investigators for determination of L_1 -, L_2 - and L_3 -subshell photoionization cross sections by measuring the intensity of L_{α} , $L_{\tau 1}$ and $L_{\tau 2}$ x-rays. In this case the $L_{\tau 1}$ and $L_{\tau 2}$ x-ray peaks do not appear separate but were resolved using spectroscopy computer software.

The coincidence technique is an older and well tried technique for selective measurements, and has been used for many of the measurements in literature [21].

Acknowledgments

Financial assistance from the Department of Science and Technology, Government of India, is gratefully acknowledged.

References

- [1] J. H. Scofield, Lawrence Livermore Laboratory Report No. UCRL-51326 (1973).
- [2] J. H. Hubbell, Wm. J. Veigele, E. A. Briggs, R. T. Brown, D. T. Cromer and R. J. Howerton, *Phys. Chem. Ref. Data* **4**, 471 (1975).
- [3] M. O. Krause, C. W. Nestor, Jr., C. J. Sparks, Jr. and E. Ricci, Oak Ridge National Laboratory Report No. 5399 (1978).
- [4] M. H. Chen, B. Crasemann and H. Mark, *Phys. Rev. A* **24**, 177 (1981).
- [5] J. H. Scofield, *Phys. Rev.* **179**, 9 (1979).
- [6] J. H. Scofield, *At. Data Nucl. Data Tables* **14**, 121 (1974).
- [7] P. V. Rao, M. H. Chen and B. Crasemann, *Phys. Rev. A* **5**, 997 (1972).
- [8] K. L. Allawadhi, Raj Mittal, B. S. Sood and N. Singh, Nuclear Science Laboratories, P. U. Patiala Report (1994).
- [9] J. H. Hubbell, P. N. Trehan, Nirmal Singh, B. Chand, D. Mehta, M. L. Garg, R. K. Garg, Surinder Singh and S. Puri, *J. Phys. Chem. Ref. Data* **23**, 339 (1994).
- [10] Raj Mittal, B. S. Sood and K. L. Allawadhi, Nuclear Science Laboratories, P. U. Patiala Report (1995).
- [11] E. J. McGurie, *Phys. Rev. A* **5**, 1043 (1972).
- [12] J. H. Hubbell, NBSIR Report No. 89-4144, US Department of Commerce, Gaithersburg, MD-20899 (1989).
- [13] K. S. Mann, N. Singh, Raj Mittal, B. S. Sood and K. L. Allawadhi, *X-Ray Spectrometry* **23**, 208 (1994).
- [14] K. S. Kahlon, K. Shatendra, K. L. Allawadhi and B. S. Sood, *Pramana* **35**, 105 (1990).
- [15] K. S. Kahlon, H. S. Aulakh, N. Singh, R. Mittal, K. L. Allawadhi and B. S. Sood, *Phys. Rev. A* **43**, 1455 (1991).
- [16] K. S. Kahlon, K. L. Allawadhi and B. S. Sood, *Pramana* **40**, 59 (1993).
- [17] K. S. Kahlon, N. Singh, Raj Mittal, B. S. Sood and K. L. Allawadhi, *Phys. Rev. A* **48**, 1701 (1993).
- [18] J. K. Sharma, Vandna, N. Singh, B. S. Sood, R. Mittal and K. L. Allawadhi, *Proc. National Symposium on Radiation Physics, Kalpakkam* (1993).
- [19] J. K. Sharma, N. Singh and K. L. Allawadhi, (in preparation).
- [20] S. L. Sorenson, S. J. Schaphorst, S. B. Whitfield, B. Crasemann and R.

- Carr, Phys. Rev. A **44**, 350 (1991).
- [21] W. Bembynek, B. Crasemann, R. W. Fink, H. A. Freund, H. Mark, C. D. Swift, R. E. Price and P. Venugopala Rao, Rev. Mod. Phys. **44**, 716 (1973).